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HIGH VOLTAGE GENERATION WITH A BETA ELECTROGENERATOR CELL

by Charles A. Low, Jr. Lewis Research Center Cleveland, Ohio

TECHNICAL PAPER proposed for presentation at Fifth Intersociety Energy Conversion Engineering Conference, ENERGY-70 Las Vegas, Neveda, September 21, 1970

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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ABSTRACT

The direct conversion of the radioactive decay energy of a beta emitter, Cerium-144, into high voltage electricity has been investigated in a coaxial cylinder cell. A 100 curie beta source absorbed in a thin graphite band on a 28 centimeter inner cylinder constituted the energy source. The energy spectrum and the output voltage were measured with a semiconductor detector. At vacuums between 1×10^{-7} and 2×10^{-4} torr, the maximum voltage was limited to 50 to 75 kilovolts, even when the electrodes were preconditioned to a higher voltage. Introduction of argon at pressures near 5×10^{-4} torr increased this voltage capability to about 200 kilovolts.

INTRODUCTION

The concept of the direct conversion into electricity of the kinetic energy associated with the charged particle emitted in radioactive decay was first suggested by Moseley¹ in 1913. However, for many years this simple nuclear cell concept was not explored; mainly, as a con-

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sequence of an insufficient supply of radioactive material. In 1952 Linder and Cristian², using a millicurie source of Strontium-90, performed an experiment which indicated an appreciable build-up of voltage from a beta source. An experiment on a cell with a 5 curie source of Polonium-210 was conducted in 1964 by Anno. ³ The results with respect to voltage build-up from the more complex alpha cell were not as encouraging as those from the earlier and simpler beta cell.

A study of Mickelsen and Low⁴ showed that a direct nuclear electrogenerator cell using a beta emitter could be fabricated as a simple light-weight space power generation system. The theoretical efficiency of coaxial cylinder beta cells has been studied for a Cerium-144 system by Cohen, ⁵ and more recently for a number of radioisotope systems by Pishunov and Favorskii. ⁶ The coaxial cylinder configuration was chosen for the present experiment, because it was practical to construct and had been evaluated well theoretically.

The object of the experimental program reported here was to study the voltage generation characteristics of a beta electrogenerator cell which had a larger physical size and a greater source strength than in previous experiments. The primary design goal was to increase the operating voltage to levels near where the efficiency is maximized. The experimental program is detailed by first describing the facility and the experimental apparatus, then by giving the techniques and procedures of the measurements, and finally by indicating the results of the experimental program.

FACILITY AND EXPERIMENTAL APPARATUS

General Facility Description

The experimental beta cell configuration, shown assembled in figure 1, is located in one quadrant of a concrete hot cell. This 2.45 meter square cell is part of the Hot Laboratory (described by Oldrieve in ref. 7) at the Plum Brook Reactor Facility of the Lewis Research Center at Sandusky, Ohio. The gamma emission at 2.18 MeV associated with the Cerium-144 decay forces remote operation of this experiment in such a hot cell.

The in-cell arrangement of the apparatus is shown in figures 2 and 3. The experimental beta cell occupies only one quadrant of the cell when assembled for testing. In the center of the cell is a remote handling machine which moves the heavy vacuum tank components; and also in conjunction with an additional unit, it is used to move the electrode components. This unit lifts these electrode components by an expansion grip which operates within the 7.5 centimeter center bore of the inner electrode sections. One of these electrode sections is the radioactive source. This is stored in the lead shield cask and is transferred to the inner electrode position when the experimental configuration is assembled for testing. The remote handling of these in-cell components required the design of much special mechanical equipment to accomplish these movements, and it is not practical to describe all this equipment in detail herein.

General Experimental Configuration

The experimental test configuration is shown assembled in figure 1, and the photograph of figure 3 shows this apparatus partially disassembled.

The vacuum tank components are stacked to form a right circular cylindrical chamber which is 183 centimeters high and 89 centimeters in diameter. This enclosure serves as the outer negative electrode or collector of the beta cell in addition to forming the vacuum chamber. The inner electrode sections stack to make a cylindrical positive electrode which is 28 centimeters in diameter and 117 centimeters long.

The chamber outer diameter is as large as is commensurate with the hot cell dimensions and handling requirements. This minimizes the electric field intensity for all voltages and any internal cell geometry. The choice of internal cell geometry is dictated by two factors. First, a voltage breakdown relationship developed by Cranberg⁸ shows that for a coaxial cylinder geometry the inner electrode diameter should be about one-third that of the outer electrode in order to minimize breakdowns. Secondly, it is desirable that the length of the electrode be such that a reasonable size region is free of end effects, so that the cell approximates the infinite coaxial cylindrical configuration on which the theoretical calculations are based. Thus, for the given outer electrode diameter the inner cell configuration represents the best design from both the standpoint of voltage breakdown and approximation to the theoretical model.

Insulator and Shorting Mechanism

The positive center electrode assembly is supported upon an insulator of polycarbonate material which is 10.2 centimeters in diameter and 61 centimeters in length. The design of the junction shielding is based on the results of Finke. ⁹ An insulator of the mechanically strong poly-

carbonate material was tested extensively at voltages up to 600 kilovolts by Koral. ¹⁰ In that program the exact geometry used in the beta cell experiment was tested and the design of the insulator and shield electrodes was optimized. The final design showed negligible current across the insulator or its electrode gap at the test voltages.

The center of the insulator is hollow and in this area a spherical tipped rod travels (fig. 1). This moving rod serves as a switch to connect the electrode assembly with an electrically isolated external connection. Thus, the current from the radioactive source at ground potential can be measured or the electrode itself can be connected to ground to short the beta cell remotely.

Radioactive Source

Cerium-144 is the choice of radioactive material for several reasons; namely, it is potentially available in large amounts, the cost per curie is low, and it is a high energy beta emitter. The latter property allows the use of a reasonably thick emitter layer.

The technique of fabrication was established in cooperation with the staff of the Isotopes Development Center at the Oak Ridge National Laboratory where the radioactive source electrode was fabricated.

The source is Cerium-144 impregnated in a porous graphite ring which is 28 centimeters in diameter, 0.159 centimeters thick, and 7.6 centimeters high. This graphite ring is mounted to the stainless steel electrode support structure. The source strength when fabricated in April 1967 was 100 curies and decayed to about 10 curies in December 1969 at the completion of this program. Figure 4 shows the decay curve

for this source for the period of the experimental testing. The graphite ring weighs 180.5 grams and contains 6.9 grams of cerium oxide of which 0.036 grams is radioactive Cerium-144. The nonradioactive cerium is added to simulate the energy absorption characteristics of a much more concentrated source layer.

Vacuum System

The vacuum chamber (i.e., the inter-electrode region) is evacuated by three 25 centimeter high speed diffusion pumps, which use a silicon operating fluid. These pumps with liquid nitrogen cooled cold baffles are located directly below the experimental chamber. The system is capable of achieving an ultimate pressure of 3×10^{-8} torr in the experimental chamber. A small line to the control area on the front side of the cell connects the vacuum system to a variable leak control valve through which argon may be introduced to increase the system pressure.

Beta Energy Detector - Mechanical

The energy of electrons emitted by the source is detected by a 5 millimeter thick lithium drifted silicon solid state detector. The detector assembly, shown in figure 1, extends from a port in the top vacuum tank section at a position opposite the radioactive band on the inner electrode. A copper cold finger serves as the mounting for the detector and provides cooling to liquid nitrogen temperatures.

Two circular apertures 2.34 millimeters in diameter spaced about 7.5 centimeters apart collimate the electrons striking the detector. The first aperture is 32 centimeters from the radioactive band on the inner electrode while the detector is located about 48 centimeters from the

source. Two lead shields are used to reduce the gamma radiation reaching the detector.

Beta Detector-Electrical

Figure 5 shows a block diagram of the electronics associated with the lithium drifted solid state energy detector. The preamplifier is located in the hot cell and is coupled to the detector with a 5 centimeter long coaxial cable. A specially designed power supply provides dc filament and other voltages to this preamplifier. These voltages with a high degree of filtering and regulation provide excellent low-noise and stability characteristics for the preamplifier system. The signal from the preamplifier is brought out of the cell to a linear amplifier located in the control console. The amplified signal is then analyzed by a 400 channel analyzer and the resulting energy spectrum is displayed on a commercial X-Y display oscilloscope. Recording of data is accomplished by photographing the spectrum display. A pulse generator is used to feed test signals to the input of the preamplifier for systems checks and calibration purposes.

External High Voltage System

A 200 kilovolt oil-filled power supply is located on the top side of the hot cell. It is used to condition the internal electrode to high voltage and also for calibration of the voltage measuring device. Figure 1 shows the cast epoxy feedthrough insulator and assembly which is used to introduce this voltage through the top of the experimental beta cell. This oil-filled feedthrough has a sliding vacuum shaft seal at the lower end. This allows the center shaft and spherical end cap to be extended downward, so that contact is made with the center electrode structure of the experimental

beta cell. After the electrode is properly conditioned, the cable and center shaft are pulled upward leaving the experimental electrode isolated from this external system.

MEASUREMENT TECHNIQUES AND PROCEDURES General Experimental Program

The sequence of experimental tests is given in this section as a guide to aid in the better understanding of the results the program discussed later. The source was fabricated in April 1967, and initial in-cell measurements began in September of 1968. At this time the source had decayed to 29 curies. These first measurements established a need for improved voltage detection accuracy. An extensive program of cooling the detector and modifying the electronics was conducted. This new system was calibrated and adjusted for optimum operation. Then a series of voltage build-up measurements at high vacuum were made culminating in a 300 hour test. Next the vacuum system was modified for the introduction of argon and tests were conducted up to a pressure of 2×10^{-4} torr. Following these tests the feedthrough for voltage conditioning of the electrodes was installed, and several measurements were made with conditioned electrodes at high The decrease of conditioning level with time was studied. Finally, measurements were made of the voltage generation at a pressure of 5×10⁻⁴ torr to complete the program.

Voltage Measurements

In this very low current beta cell a basic experimental requirement

was the measurement of voltage without loading the cell or distorting the electric field. The fact that a voltage which retards the emitted charged beta particles shifts the measured energy spectrum an amount corresponding to that voltage appeared to present a good method of measuring the voltage. Since the energy spectrum was a desired measurement, the use of the same equipment to also measure voltage was an attractive solution. The basic concept of using the spectral shift had been previously employed by Anno³ for the alpha cell. The beta particles do not have a distinct energy of emission but are emitted with a continuous spectrum of energies. Also, the range of high energy beta particles in materials is great, so that a 5 millimeter thick lithium drifted detector must be used. These two factors combined to make the concept difficult to reduce to practice for the beta cell.

Initial tests with the beta source were made using the detector at room temperature; however, these measurements indicated that the voltage range of interest was below 200 kilovolts; and thus, it was necessary to improve the resolution of the detector and to lower the electronic system noise in order to achieve accuracy. Therefore, provision was made for cooling the detector to liquid nitrogen temperatures and the electronic system noise was reduced by using a special design preamplifier power supply in cell. A 450 volt detector bias gave the best resolution for the cooled detector system. The measured resolution was about 25 kilovolts for the 626 kilovolt electron emission from a Cs-137 test source. Since the system is to be used with beta particles where the maximum energy is 3.00 MeV, the actual energy calibration was performed using a Bi-209 source which has electron

emissions up to 1.75 MeV. Simultaneously with the accumulation of the spectrum from the Bi-209 source, pulses from the pulse generator were fed into the input of the system. Thus, a calibration between the actual energy and the pulser setting was obtained. During the actual experimental testing, the calibrated test pulses from the pulse generator were used to provide the system energy calibration.

A background spectrum was measured with the primary beta source in position with the electrons blanked from the detector. This background was sufficient enough to require its subtraction from the total measured spectrum in order to accurately determine the beta spectrum itself. Since it was impossible to determine the background experimentally for every voltage measurement, a calculated correction based on this one determination was applied to the measured energy spectra in voltage measurements.

A voltage measurement was made in the following manner. With the beta cell operating at the specified test condition, an energy spectrum was accumulated with the multichannel analyzer. The analyzer accumulated the total energy spectrum from 0 to 4.00 MeV over 400 channels. A minimum of 1000 seconds was required for statistical accuracy. The usual collection time was either 3600 or 7200 seconds. The spectrum was recorded by photographing the oscilloscope display. The beta cell was shorted to ground with the switch in the insulator, and a second spectrum was collected for exactly the same time as the first and recorded. Finally, the pulser was used to calibrate the energy scale of the analyzer. The two spectra were then compared and the voltage determined from the shift after correction

for background. For this shift comparison it was sufficient to use a selected portion of 100 channels, which corresponded to energies between 1.00 and 2.00 MeV approximately.

In the latter stages of the experiment when an external source of voltage to 200 kilovolts was available, the energy shift was measured with 50, 100, 150, and 200 kilovolts applied to the beta source electrode. Agreement within 20 kilovolts was obtained at all voltages; thus, the 25 kilovolt resolution limit as measured in the initial tests appears to be a reasonable estimate of the accuracy of this method of voltage measurement.

Current Measurements

The current measurements reported in this paper were made through an electrometer and current integrator circuit which had been calibrated against a standard current source. The basic short circuit or ground current of the source was measured to ground through this instrument. Also, a 90 volt battery was used to bias the electrode both positively and negatively with respect to ground. These biases did not effect the current indicated; therefore, it can be concluded that this current measurement was actually the result of high energy particles from the source.

Vacuum Pressure Measurements

All of the vacuum system pressures were measured using a single Bayard-Alpert type ionization gauge. Since the pressure dependencies were important to this experiment, the calibration of this gauge was obtained initially by direct comparison and test on a vacuum system with

other gauges of known accuracy. At a pressure of 2×10^{-5} torr, the gauge tube and control circuit indicated a pressure that was about 20 percent low. Later a Flosdorf type McLeod gauge was connected to the experimental chamber while the system was at a pressure of 2×10^{-4} torr. This gauge indicated a pressure of twice that shown by the ion gauge. This is good agreement, since it is to be expected that the absolute pressure gauge connected to the system with a relatively long rubber hose would show a higher pressure. Thus, it can be concluded that the pressure measurements of this experiment are correct within a factor of less than two, with the actual pressure perhaps being slightly higher than that shown.

For the study of the effects of increased pressure, argon was introduced into the system through a variable leak. This leak was adjusted until the pressure was stabilized at the desired level. After one hour of operation the pressure was stable to within 10 percent even over a long duration of several days. Thus, a one hour interval was always allowed before a test measurement was taken after a pressure change.

RESULTS AND DISCUSSION

The basic measurements of this experimental program were source current and the cell voltage generation. The current measurement indicated certain physical characteristics of the source. The voltage generation was the most significant cell performance parameter, which could be measured at these source strengths with correspondingly low emission currents. The results of the current and voltage measurements with a discussion of their significance are given in the following sections.

Current Measurements

The total current emitted from the source into the cell gap as a function of the total current created by the decay process itself was used as a measure of the suitability of the source thickness and the distribution of the radioactive material. At a source strength of 29 curies the total radioactive particle emission corresponds to a total current of 17.2× 10^{-8} amp, while the measured emission current was 2.18×10^{-8} amp. Figure 4 shows that this current decreased at a rate corresponding to the decay time constant of the source. Thus, a fixed fraction of the available current was always emitted.

Emission of 12.7 percent of the maximum current through the surface was in reasonable agreement with the estimate of Cohen⁵ for this thickness of source layer. However, a thinner source would be more desirable. Nevertheless, the graphite ring approach clearly represents a suitable method of fabrication, and its use does not present any restriction on the cell operating characteristics.

Voltage Build-Up Measurements

An extensive program was conducted to evaluate the voltage generation of the experimental beta cell for two specific situations. First, the voltage generation was studied for a high vacuum inter-electrode environment. Secondly, the effect of operation with an argon gas environment in the inter-electrode gap was studied at various gas pressures.

<u>Voltage build-up at high vacuum</u>. - The results of all tests conducted at a high vacuum inter-electrode environment are given in Table I. The first test with the detector at room temperature and original commercial

electronic circuitry is shown for reference only. After improvement of the voltage measuring system to an accuracy of 25 kilovolts, the first voltage build-up tests were conducted. All voltage generation measurements were of the open circuit no-load type. These open circuit tests at high vacuum were conducted with increasing duration of elapsed time culminating in the 300 hour test shown in Table I. In all cases the voltage developed was between 50 and 75 kilovolts. The effect of time appears to be that of voltage conditioning of the electrodes since the charging time for this cell, which has a capacity of about 12 picofarads, is just a few seconds even at the lowest current levels. For the cell configuration of this experiment Koral 10 determined that the electrodes should condition at the rate of one volt per minute for the currents available from this source. Thus, for the 300 hour test a rise of 20 kilovolts above the initial threshold value should be expected. The experimental data for the 300 hour test shows a net rise between 15 and 25 kilovolts. While this is excellent agreement, the accuracy of 25 kilovolts inherent in the voltage measurements must be recognized as a serious experimental limitation on these results. A longer duration test was not feasible. A comparison of all data for unconditioned electrodes in high vacuum with that of Anno³ for the alpha cell yielded a striking similiarity in the voltage limit for natural build-up. Even though the sources emitted different particles and different geometry and materials were involved.

The fact that the voltage build-up exhibited a slight increase with time in the present experiment led to the installation of an external

power supply to more rapidly condition the electrodes. After the electrodes were conditioned to 200 kilovolts, the voltage build-up in 60 minutes was measured to be 85 or 90 kilovolts, while for a longer duration of one day a build-up of 75 kilovolts was observed. The short test was a considerable improvement over that for the unconditioned electrodes, but the longer test showed little improvement over the unconditioned situation. The 70 kilovolt level was the point at which the first currents occurred as the applied voltage was raised during the conditioning process for both Koral's work 10 and the external conditioning in these tests. Thus, it was possible that the surface conditioning was deterioating with time to the initial unconditioned level. Koral observed that the voltage conditioning level decreased with time when the electrode had no voltage applied. A similar deterioation could have been occurring for the low current beta cell. This speculation led to a test where the data was taken more rapidly as a function of time. The results of this program are shown in figure 6. The short analyzer accumulation times of 300 seconds gave less voltage accuracy than normal, but it is significant that the expected deterioration of voltage conditioning level occurred rapidly as time progressed.

The most important observation from this test is not that the voltage conditioning level decreases with time, but that the physical processes of the beta cell operation were capable of generating voltages much higher than those of the steady state limit. Thus, it was concluded that an electrode surface phenomena was responsible for this 50 to 70 kilovolt limit rather than that it was a basic property of the cell operating at high vacuum.

Therefore, it is likely that this limit can be overcome by a higher curie level source having sufficient current to condition the electrode surfaces.

Voltage build-up in argon environment. - Koral¹⁰ demonstrated that the introduction of a gas into the inter-electrode gap can greatly increase the effective electrode voltage conditioning level. In his simulation experiment, done with the same geometry electrode and insulator as in the experimental beta cell, the results showed that a factor of four gain in the effective voltage conditioning level occurred at a pressure from 7.5×10^{-5} to 2×10^{-4} torr. These pressures yield a mean free path for high energy electrons which should little effect beta cell operation in theory. The possible gain in cell voltage build-up capability made it desirable to test the beta cell in this pressure regime with argon as the background gas.

The results of these tests are shown in figure 7. The tests exhibited no improvement with increasing pressure up to 2×10^{-4} torr. It was concluded initially that the helpful effect of gas pressure was not present for the beta cell, since the effect did not occur in the pressure range anticipated from the work of Koral. ¹⁰ A similar ineffectiveness of background gas in a self generating cell was observed by Anno ³ for the alpha cell.

At the end of the experimental program the danger of destroying the heavily biased solid state voltage detector by voltage breakover due to gas pressure was an acceptable risk and further testing was done. At a pressure of 5×10^{-4} torr of argon background gas, as shown on figure 7, a dramatic build-up of voltage to the 200 kilovolt level occurred. The

bigest voltage build-up at 225 kilovolts, shown as a square of figure 7, was attributable to an electrode just conditioned to 200 kilovolts with an external supply. However, the other two measurements at 185 and 205 kilovolts were with unconditioned electrodes as were all the tests at lower pressures. At a pressure of 2×10^{-3} torr the cell is effectively shorted by the gas pressure. Thus, only a very narrow pressure range is helpful to the beta cell. This strong pressure control requirement must be considered a disadvantage to the use of a gas background; nevertheless, these results clearly show that this beneficial effect of gas pressure can be used to obtain voltage build-up to levels not achieved before in other types of cells.

An important consideration in the use of high background gas pressures is the fact that the gas environment may serve to retard or capture some of the primary beta particles before they reach the collector. As a check on the magnitude of the effect, an energy spectrum was collected both with a pressure of 5×10^{-4} torr of argon in the inter-ele electrode gap and with only a high vacuum of 1×10^{-6} torr present. No differences were discernable. Therefore, it appears that the effect of gas introduction is beneficial to beta cell operation notwithstanding the control difficulty.

SUMMARY AND CONCLUSIONS

The object of this program was to study a beta electrogenerator cell with a larger physical size and source strength than in previous experiments. A secondary goal was to improve the techniques and evaluate the materials and methods required for the construction of

such beta cells. An important contribution with respect to no-load voltage measurement was the reduction to practice for the beta cell of the measuring technique using the energy shift principle.

For the beta cell configuration and radioactive Cerium-144 source strength of this experiment, the measurements program led to four principal conclusions. First, the technique of source fabrication developed especially for this beta cell was found to be satisfactory. Second, for the source strength of this experiment, the generation of high voltages in a high vacuum environment was limited to between 50 and 75 kilovolts. Third, while the physical processes inherent in this cell and source layer configuration could produce much higher voltages, the low currents available were unable to maintain a higher level of electrode voltage conditioning. Fourth, with the introduction of argon gas as the inter-electrode environment, the voltage capability of this beta cell was increased to the 200 kilovolt level for a limited pressure range near 5×10^{-4} torr.

Based on the technology created by this experimental program and the results obtained, there appears to be no physical limitation which would prevent a beta cell with sufficient source strength from achieving much higher voltages and efficiencies in accordance with earlier theoretical predictions.

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TABLE I. - BETA CELL VOLTAGE GENERATION IN HIGH VACUUM

	Source	Elapsed	Voltage	Electrode	Pressure,	Load on
	strength,	time	generated,	surface	torr	Cell
:	curies		kilovolts	:		
Initial tests	29.0	1 hr	<100	Unconditioned 1.8×10^{-6}	1.8×10-6	None
,	15.6	$1~\mathrm{hr}$	<30	Unconditioned	1.2×10 ⁻⁷	$0.25 \times 10^{-9} \text{ amp}$
g vinger page annum	15.0	20 min	20	Unconditioned	1, 2×10 ⁻⁶	None
	14.9	20 hr	65	Unconditioned	7.0×10 ⁻⁸	None
300 Hour	13.6	$50~\mathrm{hr}$	09	Unconditioned	8.0×10 ⁻⁸	None
duration test	13.6	$100~\mathrm{hr}$	02	Unconditioned	1.2×10^{-7}	None
	13, 5	215 hr	65	Unconditioned	7.0×10^{-8}	None
	13.4	240 hr	02	Unconditioned	5.6×10^{-8}	None
	13.3	300~ m hr	22	Unconditioned	5.0×10 ⁻⁸	None
Initial tests	10°0	JUI	06	Conditioned	8.0×10^{-7}	None
with conditioning	10.0	$20~\mathrm{hr}$	75	Conditioned	1.0×10^{-7}	None

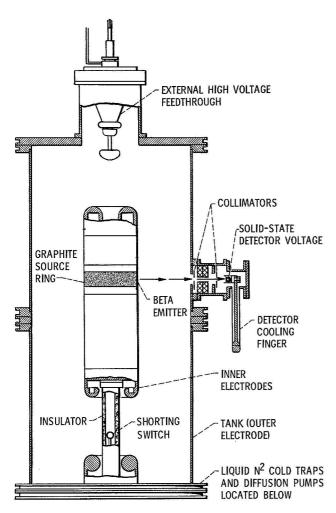


Figure 1. - Experimental beta cell configuration.

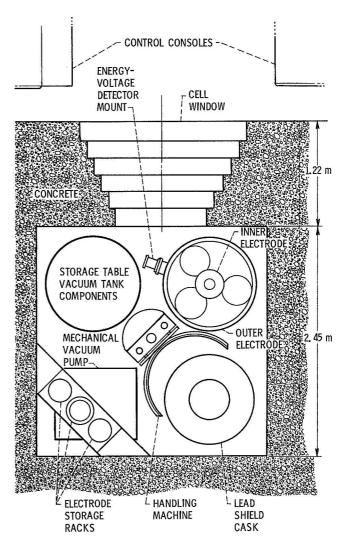


Figure 2. - General experiment layout

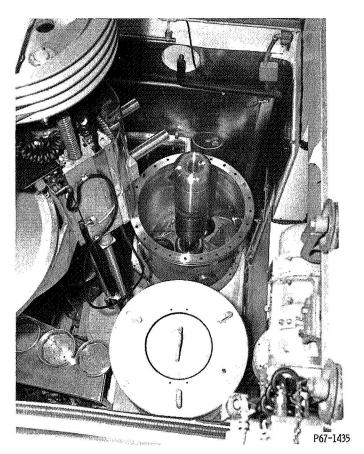


Figure 3.

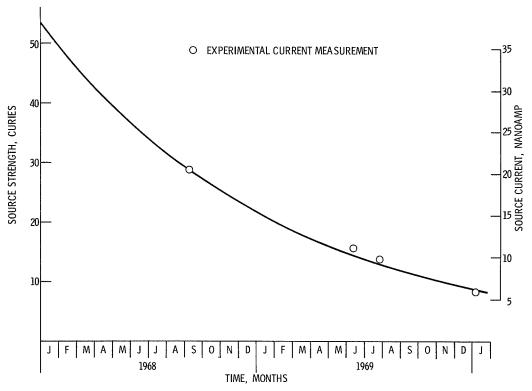


Figure 4. - Decay of source strength and emitted current with time.

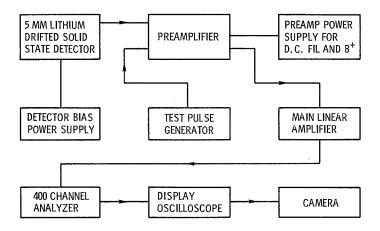
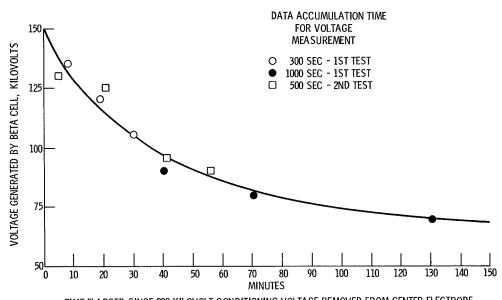


Figure 5. - Block diagram electron energy and voltage measurement system.



TIME ELAPSED SINCE 200 KILOVOLT CONDITIONING VOLTAGE REMOVED FROM CENTER ELECTRODE

Figure 6. - Loss of voltage conditioning with time.

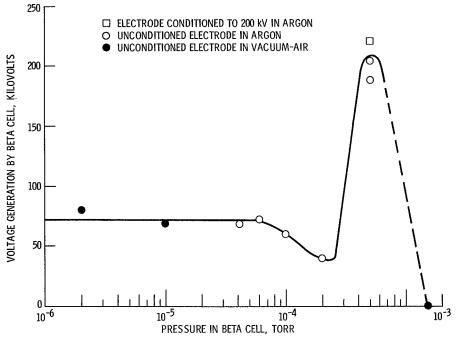


Figure 7. - Effect of pressure on voltage generation.